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Comparison of $PM_{2.5}$ carbon measurement methods in Hong Kong, China☆

Judith C. Chow[a,*], John G. Watson[a], Peter K.K. Louie[b], L.-W. Antony Chen[a], Della Sin[c]

[a]*Desert Research Institute, 2215 Raggio Parkway, Reno, NV 89512, USA*
[b]*Environmental Protection Department, 33/F, Revenue Tower, 5 Gloucester Rd., Wanchai, Hong Kong Special Administrative Region of the People's Republic of China*
[c]*Government Laboratory, 7/F, Homantin Government Offices, 88 Chung Hau St., Homantin, Hong Kong Special Administrative Region of the People's Republic of China*

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Concentrations of elemental carbon in Hong Kong are higher than in US cities, probably because of the widespread use of diesel engines.

Abstract

Samples from Hong Kong, China, were analyzed for organic carbon (OC), elemental carbon (EC), and total carbon (TC) by three thermal protocols (low-temperature IMPROVE and high-temperature STN and NIOSH) and two optical monitoring methods: reflectance and transmittance. Good agreement (±10%) for TC among the three protocols was observed for sample loadings of 1–55 μg m^{-3}. The two protocols using a reflectance pyrolysis correction showed best agreement for EC, with <20% differences found for ~80% of the samples. Hong Kong has a large diesel fleet, and for some heavily loaded samples the light transmittance was too low for quantitative detection, resulting in large uncertainties in the OC/EC split based on transmittance. Hong Kong experienced OC levels similar to those at US sites, but has much higher EC concentrations. OC/EC ratios range from 2 to 5 at two US sites and from 0.2 to 1.2 at three Hong Kong sites.

Keywords: Elemental carbon; Thermal/optical method; $PM_{2.5}$; Urban air quality; Comparison

1. Introduction

Carbonaceous material is a large component of urban particulate matter (PM) and plays an important role in urban air quality, health, visibility, and climate (Vedal, 1997; Watson, 2002; Jacobson, 2001). Carbonaceous material consists of organic carbon (OC), elemental carbon (EC), and carbonate carbon. Carbonate carbon originates from the weathering of surface soils and is negligible in most $PM_{2.5}$ (particles with aerodynamic diameter equal to or less than 2.5 μm) samples (Chow and Watson, 2002), although it is abundant in Asian dust (Cao et al., 2005). EC, also known as black carbon (BC) or soot, is the non-volatile, light-absorbing portion emitted directly into the atmosphere from combustion sources. OC contains compounds with a wide range of molecular forms and volatilities. OC and EC are commonly determined using thermal evolution methods. Different thermal methods typically give comparable

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* Corresponding author. Fax: +1 775 674 7009.
E-mail address: judy.chow@dri.edu (J.C. Chow).

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total carbon (TC) estimates but provide different partitions between OC and EC (e.g., Schmid et al., 2001; Currie et al., 2002; Fung et al., 2002).

Based on recent source tests and activity data, Streets (2003) estimates Asian anthropogenic emissions of OC and EC at 10.4 Tg $year^{-1}$ and 2.54 Tg $year^{-1}$, respectively. Emissions from China contribute 3.4 Tg $year^{-1}$ of OC and 1.05 Tg $year^{-1}$ of EC. These values exceed estimates for North America (United States and Canada) of 0.39–0.74 Tg OC and 0.31–0.59 Tg EC (Penner et al., 1993; Chen et al., 2001). These high Asian carbon emissions are reflected in ambient measurements. Dan et al. (2004) reported daily averaged $PM_{2.5}$ OC and EC of 10.7 ± 3.6 μg m^{-3} and 5.7 ± 2.9 μg m^{-3} during summer 2002 and 36.7 ± 19.4 μg m^{-3} and 15.2 ± 11.1 μg m^{-3} during winter 2002, respectively, in Beijing, China. In this case, carbon concentrations alone exceeded the United States Environmental Protection Agency (US EPA) National Ambient Air Quality Standard (NAAQS) of 15 μg m^{-3} for annual $PM_{2.5}$. Lin (2002) measured 10.5 ± 4.8 μg m^{-3} OC and 4.1 ± 1.8 μg m^{-3} EC in Kaohsiung, Taiwan, during winter. TC concentrations in urban areas of Japan approach 10 μg m^{-3} (Ma et al., 2004; Höller et al., 2001).

The Pearl River Delta (PRD) is one of the most populated areas of China and contains several large cities, heavy industries, extensive agricultural production, and major shipping ports. Hong Kong is located at the southeastern end of the PRD. With a population of 6.8 million in a ~1100 km^2 area of complex terrain (~85% of the land is covered by mountains), Hong Kong has one of the world's highest population densities. Leung (1999) reported a strong correlation between $PM_{2.5}$ mass and carbon levels. Yu et al. (2004) attributed Hong Kong PM_{10} EC to marine vessel emissions and northerly transport of air masses, with elevated carbon levels frequently occurring in winter. As part of the Hong Kong $PM_{2.5}$ Study, the first year-long fine particle measurement program in Hong Kong (Chow et al., 2002a; Louie et al., 2005a,b; Sin et al., 2005), ambient $PM_{2.5}$ was collected at three sites representing middle-scale roadside, urban-scale, and regional-scale sampling sites (Chow et al., 2002b). Organic material (OM = OC × 1.4) and EC constituted 52–75% of annual-average $PM_{2.5}$ mass at the roadside and urban sites and ~32% of $PM_{2.5}$ at the regional-scale site.

The OC/EC split in a sample depends on the thermal analysis protocol and sampling method. EC concentrations on the same sample analyzed by different methods can differ by a factor of seven (Currie et al., 2002). Chow et al. (2001, 2004), Fung et al. (2002), Yang and Yu (2002), and Chen et al. (2004) have systematically examined reasons for these discrepancies. Differences among thermal/optical analysis methods include: (1) carrier gas composition; (2) analysis temperature; (3) holding time at each temperature plateau; (4) rate of temperature ramping; (5) optical monitoring and pyrolysis correction; and (6) calibration standards. A subset of samples from the Hong Kong $PM_{2.5}$ study was analyzed with three different thermal protocols and two different optical monitoring methods at two laboratories (the Hong Kong Government Laboratory (HKGL), Homantin, Kowloon, Hong Kong, and the Desert Research Institute (DRI) Environmental Analysis Facility, Reno, NV, USA) to quantify and compare TC, OC, and EC concentrations. The objectives of this analysis are: (1) to examine the differences and ranges of OC and EC determined by different thermal/optical protocols for urban and non-urban samples, and (2) to compare ambient carbonaceous material concentrations in Hong Kong with observations from US urban/suburban areas that are based on equivalent methods.

2. Measurements

The urban-scale Tsuen Wan (TW) site, located in the New Territories, represents a highly populated residential area with mixed commercial and industrial developments. The middle-scale Mong Kok (MK) site on the Kowloon Peninsula is ~3 km southeast of TW, close to roads with heavy diesel bus, diesel taxi, and passenger car traffic; it represents a roadside microenvironment in a downtown area. The regional-scale Hok Tsui (HT) site, located at the southeast end of Hong Kong Island and ~20 km southeast of the MK site, represents a rural background/transport environment.

Twenty-four-hour (midnight to midnight) $PM_{2.5}$ samples from two collocated Partisol samplers (Rupprecht & Patashnick, Albany NY, USA) were taken every sixth day from 11/06/2000 to 10/26/2001 at the three sites (Louie et al., 2004). $PM_{2.5}$ mass and elements were measured on Teflon-membrane filters from one Partisol sampler. The second sampler was configured with a single quartz-fiber filter (#2500QAT-UP, Pall Life Sciences, Ann Arbor MI, USA) for ion and carbon analyses. The unexposed quartz-fiber filters were pre-fired at 900 °C for 3 h and acceptance tested for blank levels prior to field sampling. After sampling, the quartz-fiber samples were stored under refrigeration (<4 °C).

For samples acquired from 02/04/2001 to 10/26/2001, three punches were taken out of each 47-mm filter sample for thermal/optical carbon analysis. One of the punches was analyzed at HKGL using a Sunset Carbon Aerosol Analysis Laboratory Instrument (Sunset Laboratory Inc., Tigard, OR) following the HKGL thermal/optical transmission protocol (i.e., HKGL_TOT), while the other two punches were analyzed at DRI using a DRI/OGC carbon analyzer (Desert Research Institute, Reno, NV, USA) following the Interagency Monitoring of Protected Visual Environments (IMPROVE) thermal/optical reflectance protocol (i.e.,

IMP_TOR; Chow et al., 1993, 2001, 2004) and the DRI Model 2001 thermal/optical carbon analyzer (Atmoslytic Inc., Calabasas, CA, USA) following the Speciation Trends Network (STN; Peterson and Richards, 2002) TOT and TOR protocol (i.e., STN_TOT and STN_TOR). STN and IMPROVE are urban and non-urban $PM_{2.5}$ monitoring networks established in the United States by the EPA and the National Park Service (NPS) to acquire long-term data on $PM_{2.5}$ mass and chemical composition (Malm et al., 1994; US EPA, 1999). Field blank filters were collected and analyzed to correct ambient measurements for passive deposition and vapor adsorption (Chow et al., 1996).

Thermal/optical methods assume EC is a low-volatility carbon fraction that is not liberated in an oxygen (O_2)-free environment until temperatures of >600 °C are attained, allowing it to be separated from the more volatile OC that evolves at lower temperatures. Heating in an O_2-free environment, however, causes some OC to pyrolyze and form non-volatile, light-absorbing char that could be misclassified as EC. To ensure that the pyrolyzed or charred OC (POC) is measured as part of OC instead of EC, the reflectance or transmittance of a filter is monitored during thermal analysis, and EC is defined as the carbon fraction that evolves after the filter reflectance or transmittance returns to its initial value (the value before thermal treatment). POC is defined as the carbon measured after the introduction of a helium (He)/O_2 atmosphere but before reflectance or transmittance returns to its initial value. A negative POC is reported if reflectance or transmittance attains its initial value prior to the introduction of O_2 (i.e., early split), indicating an early combustion of EC. This may occur when EC is mixed with oxidizing minerals or catalysts in the sampled aerosol (Chow et al., 2001; Sciare et al., 2003).

The carbon analyzers used in this study operate by liberating carbon compounds under different temperature and oxidation environments, converting these compounds to carbon dioxide (CO_2) by an oxidizer (manganese dioxide [MnO_2] at 800–900 °C), reducing the CO_2 to methane (CH_4) by a methanator (nickel catalyst at ~420–500 °C in hydrogen), and then quantifying the CH_4 by a flame ionization detector (FID). The IMPROVE, STN, and HKGL analysis protocols applied in this study are compared in Table 1. To facilitate intercomparisons, the measurement results are first reported in μg per square centimeter (μg cm^{-2}) for OC and EC (1 μg cm^{-2} corresponds to an ambient concentration of ~0.5 μg m^{-3}). The minimum detection limit (MDL) for all three analytical methods is ~0.2 μg C cm^{-2}.

Table 1
Experimental parameters of the three thermal/optical analytical protocols applied in this study

Methods carrier gas	Carbon fraction	IMP_TOR, temperature (°C), time (s)[a]	STN_TOT/TOR, temperature (°C), time (s)[a]	HKGL_TOT, temperature (°C), time (s)[a]
He-purge		30, 90	30, 90	30, 90
He-1	OC1	120, 150–580[c]	310, 60	350, 70
He-2	OC2	250, 150–580	480, 60	550, 70
He-3	OC3	450, 150–580	615, 60	850, 110
He-4	OC4	550, 150–580	900, 90	Cool oven
He-5		–	Cool oven	–
O_2/He-1[b]	EC1	550, 150–580	600, 45	550, 10
O_2/He-2	EC2	700, 150–580	675, 45	600, 50
O_2/He-3	EC3	800, 150–580	750, 45	700, 40
O_2/He-4			825, 45	750, 30
O_2/He-5			920, 45	800, 30
O_2/He-6				850, 70

[a] **IMP_TOR**, thermal/optical reflectance analysis following the IMPROVE (Interagency Monitoring of Protected Visual Environments) protocol with a DRI/OGC carbon analyzer. IMP_TOR does not advance from one temperature to the next until a well-defined carbon peak has evolved (Chow et al., 1993, 2001, 2004). Filter reflectance is monitored throughout the analysis; pyrolized OC (POC) is defined as the carbon evolving between the introduction of oxygen and the return of reflectance to its initial value (the OC/EC split). POC is reported as a positive value if the OC/EC split occurs after the introduction of oxygen, and as a negative value if the OC/EC split occurs before oxygen is introduced. In either case, OC equals OC1+OC2+OC3+OC4+POC and EC equals EC1+EC2+EC3−POC. Eight well-defined fractions of carbon, including four fractions of OC (OC1, OC2, OC3, and OC4), three fractions of EC (EC1, EC2, and EC3), and POC (i.e., OP) are reported as part of the IMP_TOR protocol. **STN_TOT/TOR**, thermal/optical transmission/reflectance analysis follows the STN (Speciation Trends Network) protocol with a DRI Model 2001 thermal/optical carbon analyzer (Atmoslytic Inc., Calabasas, CA). Filter transmittance is monitored to split OC and EC in STN (STN_TOT). In this study, reflectance was also recorded during the STN analyses. The protocol that uses STN temperature plateaus but the reflectance split is referred to as STN_TOR. The STN protocol has a short and fixed residence time per temperature plateau and cannot report distinguishable carbon fractions. **HKGL_TOT**, thermal/optical transmission analysis follows the HKGL (Hong Kong Government Laboratory) protocol with a Sunset Aerosol Analysis Lab Instrument (Sunset Laboratory Inc., Tigard, OR). The HKGL transmittance protocol (HKGL_TOT) is similar to STN_TOT or NIOSH 5040) (NIOSH, 1999) but with different thermal protocols and combustion atmospheres.

[b] 2% oxygen (O_2) in helium (He) for IMPROVE and STN protocols, and 5% O_2 in He for HKGL protocol.

[c] The residence time at each temperature in the IMPROVE protocol depends on when the flame ionization detector (FID) signal returns to the baseline to achieve well-defined carbon fractions.

3. Comparisons of IMPROVE, STN, and HKGL thermal/optical carbon analyses

Results of STN and HKGL analyses are compared with those from the IMPROVE TOR (IMP_TOR) protocol, which has been applied to the US IMPROVE network since 1988; all the data is well-documented (IMPROVE, 2002; Malm et al., 1994). Table 2 summarizes the pairwise comparisons. The effective variance regression method (Watson et al., 1984) weights each data point by uncertainties of both independent and dependant variables, which can be determined from replicate analysis. This uncertainty is

Table 2
Statistical comparisons between thermal/optical reflectance (TOR) and transmittance (TOT) carbon measurements acquired in Hong Kong during 2001 (see Table 1 for experimental parameters)

Protocols		Effective variance regression[a]		Ordinary linear regression[a]		Corr. r^2	Number of pairs	Average ratio of $y/x \pm$SD	Percent deviation $\lvert y-x\rvert/x$[b]				Average (μg cm^{-2})			Student's t-test, P (sig D)[c]
x	y	Intercept ± SE (μg cm^{-2})	Regression slope ± SE	Intercept ± SE (μg cm^{-2})	Regression slope ± SE				> 10%	10–20%	20–50%	> 50%	x	y	$y-x$	
OC																
IMP_TOR	STN_TOR	0.72 ± 0.33	1.00 ± 0.04	1.49 ± 0.76	1.03 ± 0.05	0.80	129	1.16 ± 0.30	43	26	22	8	14.03	16.00	1.96	0.10
IMP_TOR	STN_TOT	0.87 ± 0.35	1.09 ± 0.04	4.47 ± 0.79	0.84 ± 0.05	0.71	129	1.24 ± 0.33	12	26	54	8	14.03	16.24	2.20	0.05
IMP_TOR	HKGL_TOT	0.32 ± 0.40	1.19 ± 0.04	−0.28 ± 0.98	1.40 ± 0.06	0.82	129	1.37 ± 0.34	13	19	47	21	14.03	19.43	5.40	0.00
STN_TOT	STN_TOR	0.11 ± 0.36	0.88 ± 0.03	−1.25 ± 0.77	1.06 ± 0.04	0.84	129	0.96 ± 0.19	36	43	18	3	16.24	16.00	−0.24	0.84
STN_TOT	HKGL_TOT	−0.30 ± 0.47	1.04 ± 0.04	−2.86 ± 1.22	1.37 ± 0.07	0.77	129	1.14 ± 0.34	57	18	13	12	16.24	19.43	3.19	0.03
STN_TOR	HKGL_TOT	−0.31 ± 0.41	1.15 ± 0.04	0.46 ± 1.07	1.19 ± 0.06	0.78	129	1.20 ± 0.30	29	25	37	9	16.00	19.43	3.44	0.02
EC																
IMP_TOR	STN_TOR	−0.29 ± 0.09	1.04 ± 0.02	1.63 ± 0.41	0.80 ± 0.01	0.97	129	0.92 ± 0.17	47	33	19	2	23.44	20.32	−3.12	0.18
IMP_TOR	STN_TOT	−6.84 ± 1.54	1.02 ± 0.04	−0.99 ± 0.57	0.90 ± 0.02	0.95	129	0.81 ± 0.16	21	28	50	1	23.44	20.07	−3.36	0.17
IMP_TOR	HKGL_TOT	−0.33 ± 0.13	0.62 ± 0.02	−0.09 ± 0.63	0.68 ± 0.02	0.90	129	0.62 ± 0.17	2	10	65	22	23.44	15.76	−7.68	0.00
STN_TOT	STN_TOR	8.37 ± 0.59	0.74 ± 0.02	3.01 ± 0.42	0.86 ± 0.02	0.96	129	1.16 ± 0.18	26	22	48	4	20.07	20.32	0.24	0.91
STN_TOT	HKGL_TOT	3.42 ± 0.93	0.62 ± 0.03	0.78 ± 0.50	0.75 ± 0.02	0.93	129	0.76 ± 0.19	21	30	40	9	20.07	15.76	−4.31	0.04
STN_TOR	HKGL_TOT	−0.37 ± 0.13	0.65 ± 0.02	−1.20 ± 0.64	0.83 ± 0.02	0.90	129	0.85 ± 1.95	5	20	60	15	20.32	15.76	−4.56	0.02
TC																
IMP_TOR	STN_TORT	0.82 ± 0.21	0.96 ± 0.01	2.14 ± 0.58	0.91 ± 0.01	0.98	129	1.02 ± 0.19	85	9	5	1	37.47	36.31	−1.16	0.73
IMP_TOR	HKGL_TOT	0.53 ± 0.19	0.90 ± 0.01	0.49 ± 0.49	0.93 ± 0.01	0.98	129	0.96 ± 0.11	76	19	5	1	37.47	35.19	−2.29	0.50
STN_TORT	HKGL_TOT	−0.07 ± 0.23	0.91 ± 0.01	−1.08 ± 0.63	1.00 ± 0.01	0.97	129	0.95 ± 0.12	80	12	7	1	36.31	35.19	−1.13	0.73

[a] Effective variance least squares (Watson et al., 1984) weights variable by precisions in both independent and dependent variables. Ordinary least squares does not weight variables by their precisions (Bevington, 1969).

[b] Percent of $y-x$ data pairs whose average differences (expressed in percent of x) fall into the specified intervals.

[c] Probability of whether protocol x and protocol y yield similar or different results. For example, a probability of 0.91 would mean there is a 91% likelihood that protocol x results are similar to protocol y results. A probability of 0.05 or less would mean that protocol x results are most likely different from protocol y results.

typically within ±10% for a measured value exceeding 10 times MDL of carbon analysis (Watson et al., 2001).

Agreement within ±10% was found for TC measured among different protocols. (TC from STN_TOT and STN_TOR are identical as they derive from the same measurement.) High correlations ($r^2 > 0.97$) and near-unity slopes (0.9–1.0) were determined for the 129 sample pairs. Student's *t*-test yields a high probability that all three TC measurements have the same mean. The average ratios of 0.95±0.12 to 1.02±0.19 also support the TC equivalence among the protocols, irrespective of different analysis temperatures and durations. The degree of carbonate decomposition may vary due to different maximum temperatures used in each protocol (e.g., calcium carbonate [$CaCO_3$], m.p: 825 °C). Two indications follow from the TC equivalence. First, the lower 800 °C maximum temperature in the IMP_TOR protocol (versus 920 °C in STN_TOT/TOR and 850 °C in HKGL_TOT) is sufficient to combust all OC and EC. Second, carbonate interferences appear to be small. Calcium (Ca) provides an upper limit estimate for carbonate if it is assumed to be entirely calcium carbonate ($CaCO_3$). Ca can also be associated with other calcium oxides and with sulfate (SO_4^{2-}) from the interaction of carbonate with sulfur dioxide (SO_2). Louie et al. (2005a) reported a mean Ca concentration of 0.10–0.17 μg m^{-3} in Hong Kong during this study period, which translates to a negligible 0.06–0.1 μg cm^{-2} of carbon as carbonate. This comparability in TC is similar to the findings of Schmid et al. (2001) and Sharma et al. (2002).

The differences in TC as a function of filter loading are plotted in Fig. 1. Samples from MK, TW, and HT have high (45–120 μg cm^{-2}), intermediate (20–45 μg cm^{-2}), and low (2–20 μg cm^{-2}) carbon loadings, respectively. For most samples, the difference is generally within ±10%. Percent differences increase when concentrations are near the MDL; therefore, 20–50% differences between STN TC and IMP TC occur for the lightly loaded samples from the regional-scale HT site. Irrespective of carbon loadings, HKGL TC is lower by 5–10% than IMP TC for >90% of samples.

Correlations (r^2) of HKGL_TOT, STN_TOT, and STN_TOR EC measurements with the IMP_TOR EC range from 0.90 to 0.97. Fig. 2, however, shows a much larger scatter in percent deviations for EC among different protocols than for TC. The closest agreement for EC was found between the STN_TOR and IMP_TOR protocols using the reflectance pyrolysis correction. The ordinary non-weighted linear regression (STN_TOR EC vs. IMP_TOR EC), which can be biased by large values, reports a slope of 0.8±0.01. When values are weighted in the effective variance regression, the slope approaches unity (1.04±0.02). For ~80% of

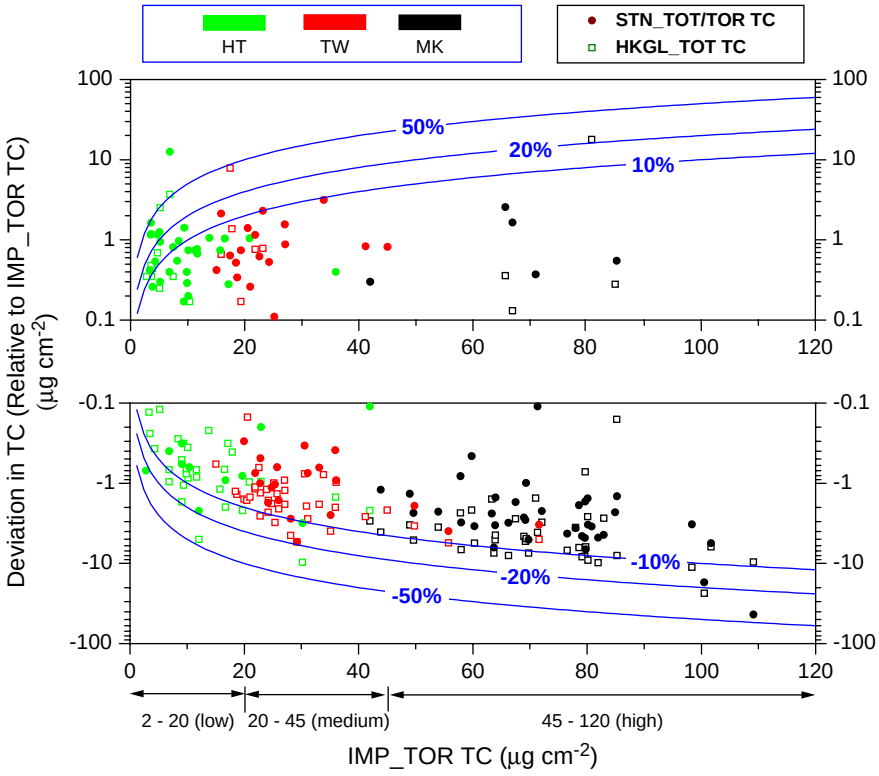

Fig. 1. Deviations in total carbon (TC) by STN_TOT/TOR and HKGL_TOT from IMP_TOR measurements acquired concurrently at Mong Kong (MK), Tsuen Wan (TW), and Hok Tsui (HT), three sites in Hong Kong, during 2001. The lines indicate the 10%, 20%, and 50% difference intervals.

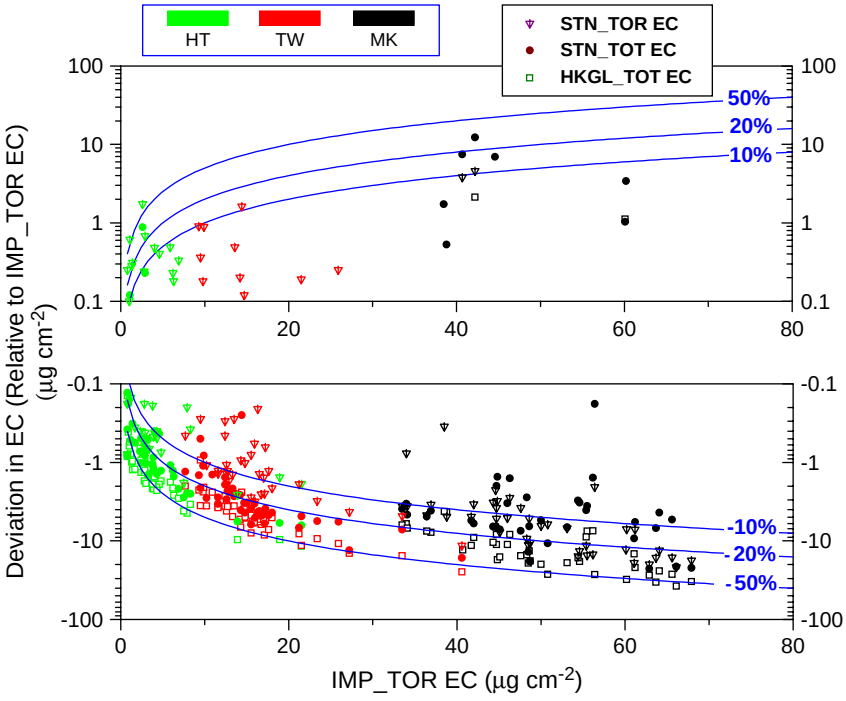

Fig. 2. Deviations in elemental carbon (EC) by STN_TOT/TOR and HKGL_TOT from IMP_TOR measurements acquired concurrently at Mong Kong (MK), Tsuen Wan (TW), and Hok Tsui (HT), three sites in Hong Kong, during 2001. The lines indicate the 10%, 20%, and 50% difference intervals.

the samples, the EC difference between the two TOR protocols is less than 20%, much better than for the other comparisons. In contrast, the largest EC difference is found between IMP_TOR and HKGL_TOT. HKGL_TOT EC is, on average, 7.7 μg cm^{-2} lower than IMP_TOR EC, and for only 12% of samples the difference between the two protocols is less than 20% (Table 2). This is consistent with Chow et al. (2004), who reported good agreement using reflectance pyrolysis corrections despite very different temperature protocols. They also found appreciable deviations between reflectance and transmission pyrolysis corrections even for the same temperature protocols. Chen et al. (2004), based on experiments and theoretical modeling, pointed out that transmittance is more influenced by pyrolyzed organic compounds from adsorbed organic vapors within the filter substrate. This partly explains the consistently lower EC determined by TOT than by TOR. EC from the STN_TOR and STN_TOT is based on the same thermal protocol but different optical correction methods. The deviation of EC between STN_TOR and STN_TOT is >20% for more than 50% of the samples. Regression slopes are 15–25% lower than unity (i.e., higher STN_TOR EC) with relatively large intercepts (3–8 μg cm^{-2}). To better understand the discrepancies in EC by TOR and TOT, the percent deviations between the protocols are plotted against the initial transmitted laser signal in Fig. 3. It appears that STN_TOR EC is 10–40% higher than STN_TOT EC except for heavily loaded (dark) samples in which the initial transmitted signal is less than 10 counts (i.e., MDL of transmittance detector). For such dark samples, the initial transmittance through a filter is undetectable by the instrument, causing large uncertainties in the OC/EC split that relies on detecting the transmittance baseline. Earlier-than-actual OC/EC splits by transmittance were triggered by random signal fluctuations in most (if not all) dark samples from MK. Irrespective of the darkness of the sample, initial filter reflectance is almost always detectable (i.e., exceeding the optical detection limit). Extra caution should be exercised when the transmittance pyrolysis correction is applied to heavily loaded samples that are essentially opaque.

EC from the HKGL_TOT and STN_TOT methods was determined by similar thermal protocols and by the transmittance pyrolysis correction. The different designs of the Sunset and DRI instruments, however, may contribute to differences in EC levels. For samples with light-to-intermediate loadings, HKGL_TOT measures the lowest EC with negative percent differences with respect to STN_TOT EC (see Fig. 3). The slope (0.62–0.75) is 25–40% lower than unity. Similar to the STN_TOT/TOR EC comparison, the percent deviation between HKGL_TOT and STN_TOT EC is larger than 20% for ~50% of the samples. For heavily loaded samples, both TOT methods are affected by undetectable transmittance.

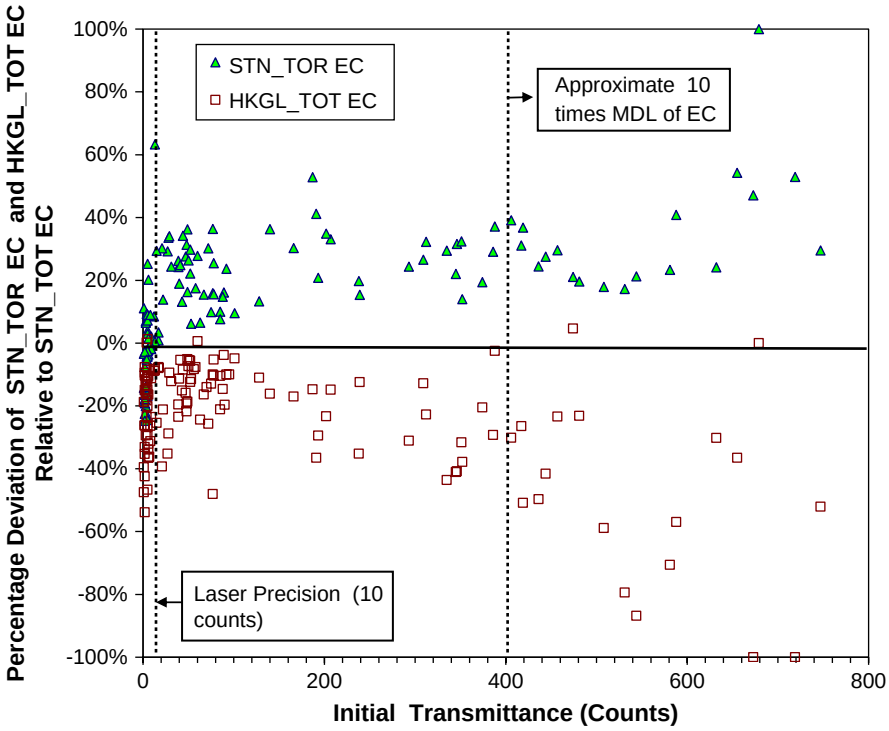

Fig. 3. Percent deviation of elemental carbon (EC) by STN_TOR and HKGL_TOT from STN_TOT as a function of initial filter transmittance.

OC is complementary to EC by definition (i.e., OC = TC − EC). The paired statistical comparisons among the four OC measurements are shown in Table 2. Absolute differences in OC ranged from −0.24 μg cm^{-2} between STN_TOT and STN_TOR to 5.4 μg cm^{-2} between IMP_TOR and HKGL_TOT. OC by STN_TOT and STN_TOR correlated best ($r^2 = 0.84$) with <50% variations in 97% of the samples. Similar percent deviations were found between OC by IMP_TOR and STN_TOT, with an average difference of 2.2 μg cm^{-2} and a lower correlation ($r^2 = 0.71$). The generally weaker correlations between OC measurements, in comparison with EC, are likely due to the lower average OC concentration across the three sampling sites. These averages largely reflect the composition of heavily loaded samples from MK, in which EC is >50% higher than OC.

4. Ambient $PM_{2.5}$ OC and EC concentrations

Ambient $PM_{2.5}$ concentrations were calculated based on sampling flow rate, duration, and deposit area. At the TW site, $PM_{2.5}$ TC concentrations of 13.9 ± 5.5 μg m^{-3} found in this study were comparable to the annual average PM_{10} TC concentrations of 13.3–17.0 μg m^{-3} (HKGL_TOT protocol) reported by Yu et al. (2004) for the 1998–2001 period. Differences were found at the roadside MK site with $PM_{2.5}$ TC concentrations of 36.1 ± 7.4 μg m^{-3} in this study, compared with PM_{10} TC concentrations of 17.5–26.8 μg m^{-3} by Yu et al. (2004). PM_{10} OC varied by a factor of 1.5 from 1998 to 2001, but smaller year-to-year variations (~20–40%) were found for EC (Yu et al., 2004). Due to the known differences in the OC/EC split, it is difficult to compare Yu et al. (2004) with the current study.

The OC/EC ratio often reflects the type of primary pollution sources as well as the contribution of secondary organic aerosol. Average ambient concentrations of OC, EC, and TC, and their carbon fractions following the IMPROVE protocol, are shown by site in Table 3. Fig. 4 compares the Hong Kong OC/EC relationships with 24-h measurements from Fort Meade (FME), MD, and Fresno, CA, USA. The sampling and analysis techniques as well as data processing/validation procedures for the FME and Fresno sites were similar to those in this study. FME is located in an open field near the Baltimore/Washington corridor (Chen et al., 2001, 2002, 2003) and ~100 m from the nearest minor roads and buildings. Several major highways (MD-295, MD-175, and MD-32), Interstate 95 (I-95), and US Route 1 are 1–6 km from the sampling site. The Fresno site is one of the US EPA Supersites, located near downtown Fresno, a major city (population ~500,000) in California's San Joaquin Valley (Watson et al., 2000; Watson and Chow, 2002a,b). It is located in an urban/commercial/residential area ~1 km from an 8-lane state highway and is impacted by residential

Table 3
Average ambient OC, EC, and TC concentration at three sites in Hong Kong following IMPROVE protocol (IMP_TOR)

	Middle-scale roadside	Urban-scale		Regional-scale	
	Mong Kok (HK)[a] (2/04/2001–10/26/2001)[f]	Fresno, CA (USA)[b] (11/15/2001–12/12/2002)[f]	Tsuen Wan (HK)[c] (2/04/2001–10/26/2001)[f]	Hok Tsui (HK)[d] (2/04/2001–10/26/2001)[f]	Fort Meade, MD (USA)[e] (12/31/1999–8/01/2000)[f]
OC ($\mu g\ m^{-3}$)	10.75±4.11	11.45±9.69	5.56±2.64	3.14±2.25	2.73±1.31
EC ($\mu g\ m^{-3}$)	24.85±4.82	2.82±2.23	7.89±3.12	2.21±2.2	0.93±0.49
TC ($\mu g\ m^{-3}$)	36.06±7.39	14.26±11.83	13.91±5.48	5.82±4.3	3.66±1.77
OC1 ($\mu g\ m^{-3}$)	2.03±1.03	1.53±1.78	0.64±0.46	0.22±0.24	0.25±0.19
OC2 ($\mu g\ m^{-3}$)	2.42±0.92	2.02±1.64	1.38±0.57	0.68±0.47	0.63±0.31
OC3 ($\mu g\ m^{-3}$)	4.01±1.99	5.26±3.87	2.41±1.55	1.26±1.21	0.68±0.36
OC4 ($\mu g\ m^{-3}$)	6.11±4.72	3.08±2.99	3.23±2.7	1.4±1.62	0.56±0.33
POC ($\mu g\ m^{-3}$)	−3.81±5.06	−0.44±1.07	−2.11±2.6	−0.42±1.39	0.58±0.32
EC1 ($\mu g\ m^{-3}$)	20.52±4.6	2.21±1.82	5.28±1.37	1.55±1.09	0.95±0.68
EC2 ($\mu g\ m^{-3}$)	0.34±0.46	0.23±0.15	0.34±0.26	0.24±0.09	0.51±0.24
EC3 ($\mu g\ m^{-3}$)	0±0.02	0.02±0.03	0±0.02	0±0.02	0.04±0.04

Data are compared to IMP_TOR measurements acquired from Fort Meade, Maryland (FME), and Fresno, California, USA.

[a] Middle-scale roadside site on the Kowloon Peninsula, Hong Kong.

[b] Urban-scale commercial/residential site in central California, USA.

[c] Urban-scale commercial/residential site in the New Territories, Hong Kong.

[d] Regional-scale rural background/transport site located at the southeast end of Hong Kong Island and ~20 km southeast of the Mong Kong (MK) site.

[e] Regional-scale suburban site located in the middle of the Baltimore/Washington corridor.

[f] Number of samples in the comparison are 43 from Hong Kong taken on an every-sixth-day sampling schedule, 60 from Fresno, CA, taken on an every-sixth-day sampling schedule, and 97 from Fort Meade, MD, taken on a daily sampling schedule during January, April, and July 2000.

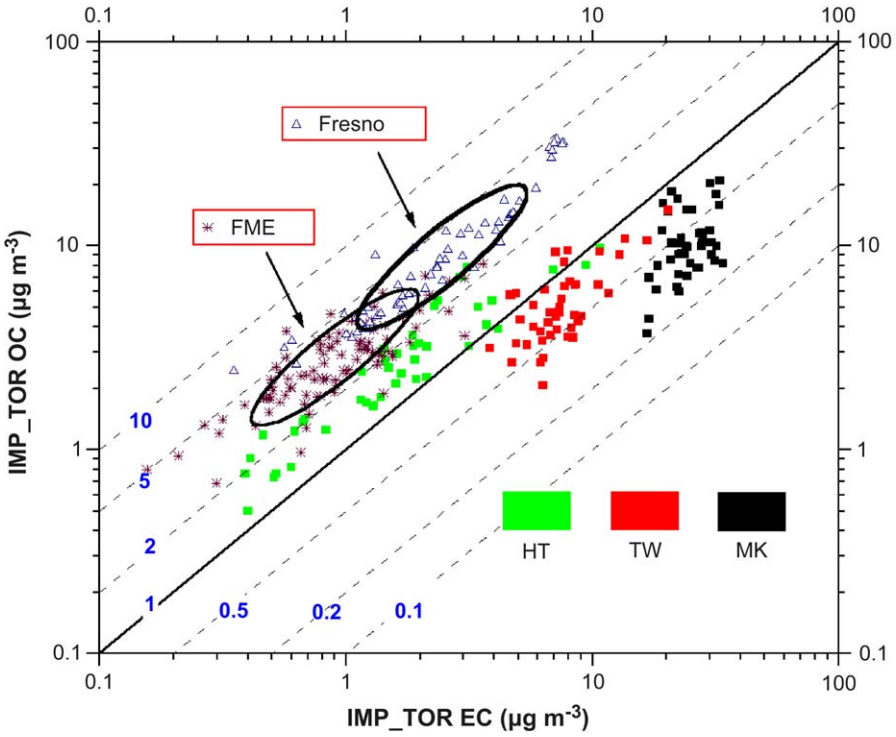

Fig. 4. Ambient OC and EC concentrations acquired at the middle-scale roadside Mong Kong (MK), urban-scale Tsuen Wan (TW), and regional-scale Hok Tsui (HT)—three sites in Hong Kong—during 2001, compared to measurements from the urban-scale Fresno site in California, USA, and the regional-scale Fort Meade (FME) site in Maryland, USA. The dotted lines indicate OC/EC ratios.

wood combustion during winter, in addition to vehicle exhaust and cooking emissions.

OC concentrations at the HT site (0.3–7 μg m^{-3}) are similar to OC levels at the suburban FME site. OC concentrations are higher at the TW and MK sites (2–20 μg m^{-3}) and similar to those at the urban Fresno site. The OC/EC ratio is between 2 and 5 at both the FME and Fresno sites, but it is much lower (0.2–3) in Hong Kong. At the roadside MK site, where particulate carbon is dominated by on-road mobile emissions, EC concentrations always exceed 16 μg m^{-3} and OC/EC ratios are between 0.2 and 1.0. Among all potential sources (e.g., gasoline-, compressed-natural-gas-fueled and diesel-fueled vehicle exhaust; cooking; vegetative burning; and biogenic), diesel emissions are most likely to produce such low OC/EC ratios. Diesel vehicles accounted for >60% of the total vehicle miles traveled (VMT) in Hong Kong during the study period. Watson et al. (1994) reported OC/EC ratios of 1.2 in diesel exhaust and 2.2 in gasoline exhaust. The OC/EC ratio was higher (1–3) at the regional-scale HT site, consistent with contributions from other sources. Although on-road mobile sources likely dominate particulate carbon emissions around FME as well, diesel vehicles account for only 4–7% of the total VMT in Maryland (Maryland Department of the Environment, 1999). At Fresno, cooking and wood burning can be as important as vehicle emissions (Schauer and Cass, 2000; Watson and Chow, 2002b), and they are usually associated with higher OC/EC ratios in ambient PM.

Temperature-resolved carbon fractions reported by the IMPROVE protocol (Table 1) could be useful for source apportionment of carbonaceous aerosol. Watson et al. (1994) demonstrated appreciable differences between the carbon fraction abundances in diesel soot and in gasoline vehicle exhaust, and they have been included in recent receptor modeling studies (e.g., Kim and Hopke, 2004; Kim et al., 2004). The carbon fraction measurements, however, appear to be more sensitive to subtle analytical conditions than OC and EC measurements, and are usually associated with higher uncertainties. Table 3 shows that low-temperature OC (OC1 at 120 °C) is more abundant at the roadside MK and urban Fresno sites, consistent with liquid fuel in fresh emissions from engine exhaust during frequent stops and accelerations. Negative POC (reflectance returns to baseline before the introduction of O_2 during thermal analysis) is found most frequently at the roadside MK site, partially due to the uncertainties in optical monitoring on very dark, heavily loaded samples. At the roadside and urban sites, most of the EC is found in the low-temperature EC1 (at 550 °C) fraction. Ratios of high-temperature EC2 (at 700 °C) to low-temperature EC1 (at 550 °C) are higher at regional-scale sites than at middle-scale roadside and urban-scale sites.

5. Conclusions

Fine particulate TC and carbon fractions were determined using four thermal/optical methods for a year-long ambient $PM_{2.5}$ study in Hong Kong, China. Though measured by different laboratories with different instruments and analytical protocols, TC data from the IMP_TOR, STN_TOT/TOR, and HKGL_TOT protocols show agreement within ~10% for measured concentrations higher than 10 times the MDL. Deviations in OC and EC measurements are more substantial. Best agreement in EC (<20% differences for ~80% of samples) was observed between IMP_TOR and STN_TOR when reflectance was used for pyrolysis correction. The poorest agreement in EC (<20% differences for ~12% of samples) was found between IMP_TOR and HKGL_TOT, with an average difference (IMP_TOR minus HKGL_TOT) of 7.7 μg cm^{-2}. STN_TOT EC was generally 10–40% lower than STN_TOR EC except for heavily loaded samples. The transmittance pyrolysis correction was most affected by heavily loaded samples because the transmitted laser signal was too low to be detected.

Hong Kong's ambient $PM_{2.5}$ contains substantial carbonaceous material that could impact the regional air quality. TC (overall mean: 37.5 μg m^{-3}) reached a level similar to that in Beijing, China, during winter, and it varied according to sampling sites. At the urban TW site, TC concentrations were similar to those at the urban Fresno, CA, site but EC concentrations were 2–3 times higher. At the regional-scale HT site, OC and EC concentrations were much lower, but still appreciably higher than those at the regional-scale Fort Meade, MD, site. The OC/EC ratio differed by more than tenfold, from 2–5 at the two US sites to 0.2–1.2 at the Hong Kong roadside and urban sites. The very low OC/EC ratios of 0.2–1.0 at the roadside MK site are consistent with a strong influence of on-road diesel emissions.

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